

Clustering of phosphorus atoms in silicon

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Abstract

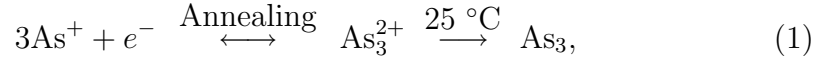
To explain the effect of electron density saturation at high phosphorus concentrations the model of negatively charged phosphorus clusters was compared with experimental data. A number of negatively charged clusters incorporating a point defect and phosphorus atoms $(DP_1)^-$, $(DP_2)^-$, $(DP_2)^{2-}$, $(DP_3)^-$, $(DP_3)^{2-}$, $(DP_4)^-$, and $(DP_4)^{2-}$ were investigated at temperatures of 920 and 1000 °C. It was established that for clusters incorporating more than one phosphorus atom the calculated electron density reached a maximum value and then decreased monotonically and slowly with an increase in the total dopant concentration. If a cluster incorporates 3-4 phosphorus atoms, the calculated dependencies of carrier concentration agree well with the experimental data for both temperatures regardless of the negative charge state of the cluster (– or 2–). Moreover, for all doubly negatively charged clusters investigated the fitting parameter is independent of the temperature. It means that formation of doubly negatively charged clusters is most likely but it is difficult to choose between clusters incorporating two, three or four phosphorus atoms. From the good agreement of the calculated curves with the experiments it follows that the model based on the formation of negatively charged clusters can be used in simulation of high concentration phosphorus diffusion.

Keywords: clusters; diffusion; doping effects; phosphorus; silicon

1 Introduction

At the present time, low energy high fluence ion implantation is widely used for producing the active regions of modern integrated microcircuits. A combination of ion implantation having energies of about 10 keV with rapid thermal annealing allows one to create doped layers with a depth of about 100

nm [1, 2]. To calculate the distributions of electrically active dopant atoms in such small regions, models of clustering having a high degree of accuracy should be used. The implementation of the adequate models of clustering is very important because saturation of electron density is observed experimentally for high dopant concentrations (see, for example [3]). Thus, only a part of dopant atoms is electrically active, and we have difficulties in achieving high carrier concentrations. Unfortunately, all up-to-date models cannot adequately explain this phenomenon. Really, it is commonly accepted that the model of Tsai *et al.* [4] was the first to account for saturation of charge carriers at an increasing doping level [5]. In the model of [4] it is supposed that arsenic clustering occurs as a result of the reaction



where As^+ is the substitutionally dissolved arsenic atom participating in cluster formation; e^- is the electron.

The main feature of the model of [4] is the assumption that clustered As atoms are electrically active (positively charged) at annealing temperatures, while at room temperature they are neutral. Therefore, the electron concentration at room temperature n_R is approximately equal to the concentration of substitutionally dissolved arsenic atoms C . The assumption that As clusters are capable of trapping the conduction electrons and becoming neutral during cooling allows one to explain the saturation of electron density when $C^T \rightarrow \infty$ [4, 5]. Here $C^T = C + C^{AC}$ is the total concentration of dopant atoms; C^{AC} is the concentration of dopant atoms incorporated into clusters. In Fig. 1, the electron density at room temperature n_R calculated according to the model of Tsai *et al.* [4] is compared with the experimental data obtained by Nobili *et al.* [6]. The value of the fitting parameter in the mass action law for reaction (1) was chosen to satisfy the experimentally observed value of electron density saturation n_{max} which was equal to $2.11 \times 10^8 \mu\text{m}^{-3}$ [3].

As can be seen from Fig. 1, the curve calculated disagrees with the experimental dependence of electron density on the total dopant concentration. In Fig. 1, the dependences $n \approx n_R$ calculated for the neutral clusters VAs_2 and VAs_4 are also presented. Here $n = n(C^T)$ is the electron concentration at the temperature of cluster formation. From the calculations presented in Fig. 1 it follows that when neutral clusters are formed, the electron density increases monotonically and saturation is not observed. Thus, dependences of electron

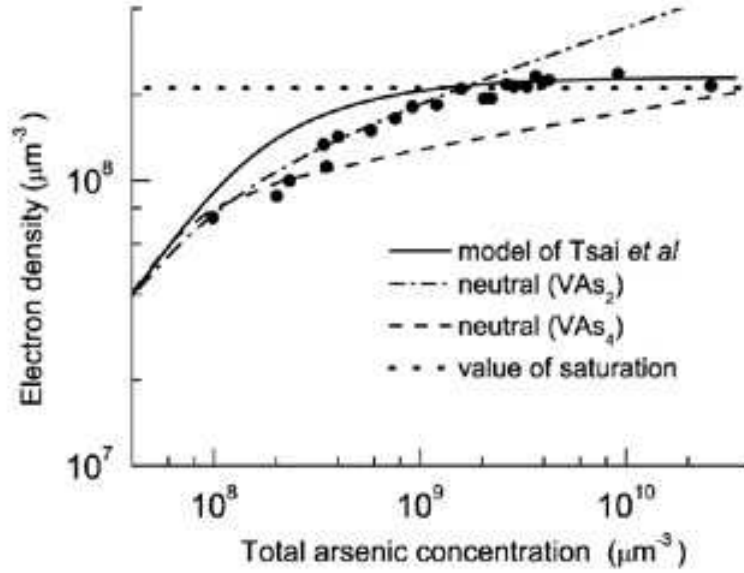
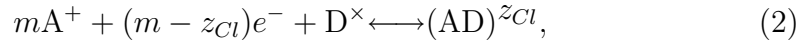


Figure 1: Calculated electron concentrations vs. the total arsenic concentration for different models of clustering: solid line - model of Tsai *et al.* (Ref. [4]), dash-dotted line - neutral VAs_2 clusters, and dashed line - neutral VAs_4 clusters. The experimental data (circles) are taken from Nobili *et al.* [6] for diffusion at a temperature of 900 °C. Dotted line represents the value of saturation.

density calculated for neutral clusters also disagree with experimental data.

In [7], the clustering phenomenon was investigated with the example of Ge doped by As. It was proposed that cluster formation occurred due to the reaction



where A^+ is the substitutionally dissolved donor atom; D^\times is the intrinsic point defect in the neutral charge state; m is the number of substitutionally dissolved atoms participating in the reaction; $(AD)^{z_{Cl}}$ is the cluster formed; z_{Cl} is the charge of this cluster.

It is supposed that a cluster has the same charge during annealing and at room temperature. Based on this assumption, the dependence between the total dopant concentration C^T and n_R was obtained in [7] from the mass

action law for reaction (2)

$$C^T - n_R = K(T) \frac{n_R^{(m - z_{Cl})} (m n_R - z_{Cl} C^T)^m}{(m - z_{Cl})^m - 1}, \quad (3)$$

where $K(T)$ is the constant of local equilibrium for reaction (2).

Two different cases of cluster formation were investigated analytically based on dependence (3): (i) formation of neutral clusters ($z_{Cl}=0$) and (ii) formation of negatively charged clusters including one dopant atom and a lattice defect. It was found that for neutral clusters the dependence $n_R = n_R(C^T)$ has the following form:

$$\lg n_R = -\frac{1}{2m} \lg(mK) + \frac{1}{2m} \lg C^T. \quad (4)$$

It follows from expression (4) that at high impurity concentrations the line with the angle $\frac{1}{2m}$ in the logarithmical coordinates is asymptotic for the dependence $n_R = n_R(C^T)$ [7]. This analytical result agrees with the numerical solutions presented in Fig. 1.

In the case of a negatively charged cluster which incorporates one impurity atom and a lattice defect Eq.(3) yields

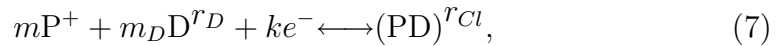
$$C^T = n_R \frac{1 + K(T) n_R^{(1 + |z_{Cl}|)}}{1 - |z_{Cl}| K(T) n_R^{(1 + |z_{Cl}|)}}. \quad (5)$$

It follows from Eq.(5) that saturation of electron density occurs if the dopant concentration approaches infinity. The maximum electron concentration n_{Rmax} has the following value [7] :

$$n_{Rmax} = [|z_{Cl}| K(T)]^{-\frac{1}{1 + |z_{Cl}|}}. \quad (6)$$

Thus, the model of [7] was the first to account for saturation of charge carriers at an increasing doping level.

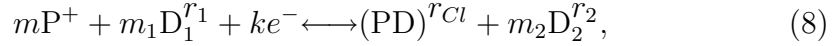
In the paper by Velichko *et al.* [8], the clustering of phosphorus atoms was investigated numerically. Due to numerical solution, the limitations of [7] were removed. It was proposed that the formation of negatively charged phosphorus clusters occurred due to the reaction



and that the charge of a phosphorus cluster does not change during cooling. Here P^+ is the substitutionally dissolved phosphorus atom; D is the point defect participating in clustering; $(PD)^{r_{Cl}}$ is the cluster formed; m , m_D , and k are the numbers of impurity atoms, defects, and electrons participating in the cluster formation, respectively; r_D and r_{Cl} are the charge states of the defect and cluster, respectively.

The possibility of formation of negatively charged phosphorus clusters is indirectly confirmed in [9, 10]. It follows from calculations [9, 10] that singly $(VAs_3)^-$ and doubly $(VAs_2)^{2-}$ negatively charged clusters can be formed at high arsenic concentrations.

The reaction used in [8] can be generalized to take into account the possibility of defect generation during clustering:



where $D_1^{r_1}$ and $D_2^{r_2}$ are the defects providing cluster formation and being generated during clustering, respectively; m_1 and m_2 are the numbers of the defects participating in reaction (7); r_1 and r_2 are the charge states of these defects, respectively. For example, $D_2^{r_2}$ can be the self-interstitial generated during clustering [11].

The charge conservation law is valid for chemical reaction (8). This conservation law is of the form

$$m + m_1z_1 - k = z_{Cl} + m_2z_2, \quad (9)$$

where z_1 and z_2 are the charges of defects $D_1^{r_1}$ and $D_2^{r_2}$, respectively.

For reaction (8) and conservation law (9), the equations obtained in [8] to describe clustering are modified to the following form:

$$C^{AC} = K \tilde{C}_D \chi^{(-z_1 m_1 + k + z_2 m_2)} C^m, \quad (10)$$

$$\begin{aligned} \chi = & \frac{1}{2n_i} \left[C + \frac{z_{Cl}}{m} C^{AC} - C^B + \right. \\ & \left. + \sqrt{\left(C + \frac{z_{Cl}}{m} C^{AC} - C^B \right)^2 + 4n_i^2} \right], \end{aligned} \quad (11)$$

where

$$K = \frac{m K_L (h_{D1}^{r_1})^{m_1} n_i^{(-z_1 m_1 + k + z_2 m_2)} (C_{eq}^{D1\times})^{m_1}}{K_R (h_{D2}^{r_2})^{m_2} (C_{eq}^{D2\times})^{m_2}}, \quad (12)$$

$$\tilde{C}_D = \frac{(\tilde{C}_{D1})^{m_1}}{(\tilde{C}_{D2})^{m_2}}, \quad \tilde{C}_{D1} = \frac{C^{D1\times}}{C_{eq}^{D1\times}}, \quad \tilde{C}_{D2} = \frac{C^{D2\times}}{C_{eq}^{D2\times}}. \quad (13)$$

Here χ is the electron density normalized to the concentration of intrinsic charge carriers in a semiconductor during diffusion n_i ; C^{AC} and C^B are the concentration of clustered phosphorus atoms and total concentration of acceptors, respectively. The parameter K has a constant value depending on the temperature of diffusion. This value can be extracted from the best fit to the experimental data. The parameters $h_{D1}^{r_1}$ and $h_{D2}^{r_2}$ are the constants for the local thermodynamic equilibrium in the reactions when neutral defects D_1^\times and D_2^\times are converted into charge states r_1 and r_2 , respectively; $C^{D1\times}$ and $C^{D2\times}$ are the concentrations of defects D_1^\times and D_2^\times , respectively. The quantities $C_{eq}^{D1\times}$ and $C_{eq}^{D2\times}$ represent the equilibrium concentrations of these defects in the neutral charge state.

In [8], the formation of singly negatively charged clusters incorporating one phosphorus atom or two phosphorus atoms was investigated. It was shown that in the first case the concentration of charge carriers reached saturation if the concentration of substitutionally dissolved dopant atoms and correspondingly, the total concentration of phosphorus atoms had increased. Thus, these calculations agree with predictions of [7]. In the second case, the electron density reached a maximal value and then monotonically decreased. Unfortunately, in this paper a detailed comparison of calculated distributions of electron density with experimental data was not carried out. Nor other cluster species were considered. Therefore, the purpose of this study is to investigate systematically the formation of different types of phosphorus clusters and compare the calculated dependences of electron density with experimental data.

2 Calculations

The system of equations (10),(11) obtained in [8] and modified in this paper allows one to consider different types of negatively charged clusters. In the

present investigation the electron density $n_R \approx n = n(C^T)$, concentration of substitutionally dissolved phosphorus atoms $C = C(C^T)$, and the concentration of clustered phosphorus atoms $C^{AC} = C^{AC}(C^T)$ depending on the total phosphorus concentration C^T were calculated for the clusters $(VP_1)^-$, $(VP_2)^-$, $(VP_2)^{2-}$, $(VP_3)^-$, $(VP_3)^{2-}$, $(VP_4)^-$, and $(VP_4)^{2-}$. To calculate these functions, a numerical solution of the system of nonlinear equations (10), (11) was obtained by Newton's method.

For comparison with the experimental data of [12] and [13], the processing temperature was chosen to be 920 °C ($n_i = 5.684 \times 10^6 \mu\text{m}^{-3}$) and 1000 °C ($n_i = 8.846 \times 10^6 \mu\text{m}^{-3}$), respectively. In the experiments carried out in [12], the gas composition containing 0.27 % POCl_3 was used as a source. The diffusion was carried out in a low dislocation density, (111) oriented, boron doped, Czochralsky pulled silicon with a resistivity of 1 $\Omega \text{ cm}$. Duration of diffusion was 7 minutes at a temperature of 920 °C. The distribution of carriers concentration was extracted using the incremental sheet resistance and Hall measurements, performed after thinning of the specimens by anodic oxidation and oxide stripping. Neutron activation analysis was used for measuring the total concentration of impurity atoms. In [13], silicon wafers, (100) oriented, p-type of 10 $\Omega \text{ cm}$ resistivity, were heavily implanted with phosphorus at a fluence of $1.0 \times 10^{17} \text{ cm}^{-2}$ and an energy of 100 keV and then with a fluence of $5.0 \times 10^{16} \text{ cm}^{-2}$ at 50 keV. The samples were furnace annealed at a constant temperature of 1000 °C for 15 min. Secondary neutral mass spectroscopy (SNMS) was employed to measure the dopant concentration profiles of the samples after annealing. Carrier concentration and mobility profiles were determined by the accurate incremental sheet resistance and Hall measurements, performed after thinning of the specimens by anodic oxidation and oxide stripping.

In Fig. 2, the electron density calculated for the formation of $(DP_1)^-$ clusters is presented. As can be seen from Fig. 2, the saturation of electron density occurs at extremely high values of the total impurity concentration C^T and calculated dependence $n_R \approx n = n(C^T)$ disagrees with the experimental data. The disagreement is much less but also takes place for the formation of the $(DP_2)^-$ clusters.

The electron densities calculated for the clusters $(DP_2)^{2-}$, $(DP_3)^{2-}$, $(DP_3)^-$, and $(DP_4)^-$ are presented in Figs. 3a, 3b, 4a, and 4b, respectively. As can be seen from these figures, a good agreement is observed between the calculated curves and experimental data especially for the case of $(DP_4)^-$ cluster formation. As follows from the calculated curves, for the clusters in-

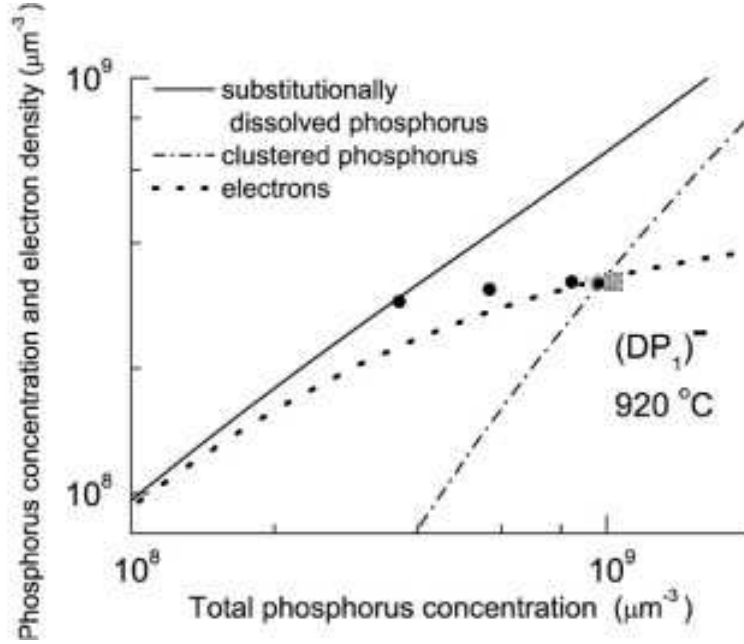
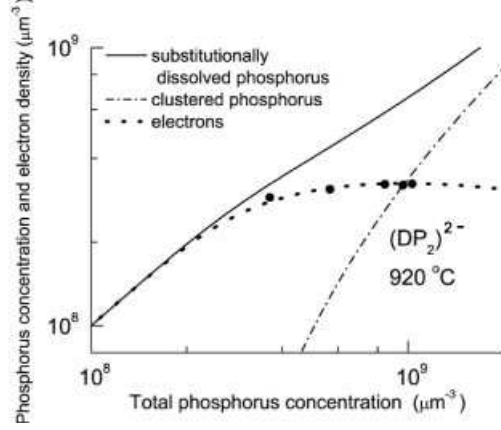


Figure 2: Calculated concentrations of substitutionally dissolved phosphorus atoms (solid line), clustered phosphorus atoms (dash-dotted line), and electron density (dotted line) vs. the total dopant concentration when singly negatively charged clusters incorporating a point defect and one phosphorus atom are formed. The experimental data (circles) are taken from Masetti *et al.* [12] for diffusion at a temperature of 920 °C.

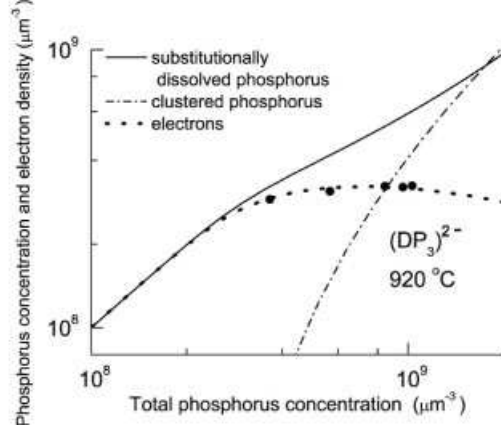
incorporating more than one phosphorus atom the electron density reaches a maximum value and then decreases monotonically and slowly with increasing C^T .

To calculate the electron densities presented, the generalized system of equations (10),(11) was transformed due to assignment of the cluster charge state, a number of defects participating in cluster formation (one in the cases under consideration), and a number of phosphorus atoms incorporated into the cluster. For example, the following equations describing the concentration of phosphorus atoms incorporated into the $(DP_4)^-$ clusters and electron density corresponding to the formation of these clusters can be obtained from the system (10),(11):

$$C^{AC} = K \tilde{C}_D \chi^5 C^4, \quad (14)$$



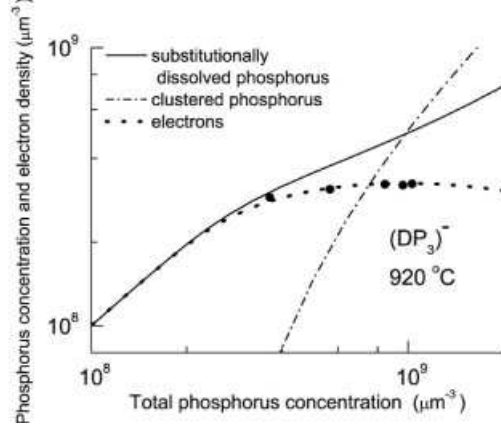
(a)



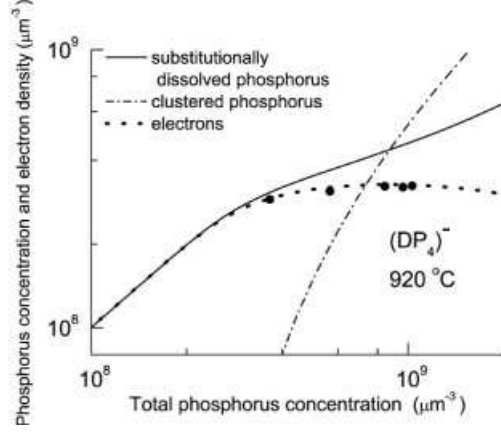
(b)

Figure 3: Calculated concentrations of substitutionally dissolved phosphorus atoms (solid line), clustered phosphorus atoms (dash-dotted line), and electron density (dotted line) vs. the total dopant concentration for the formation of doubly negatively charged clusters incorporating two (a) and three (b) phosphorus atoms. The experimental data are taken from Masetti *et al.* [12] for diffusion at 920 °C.

$$\chi = \frac{1}{2n_i} \left[C - \frac{1}{4} K \tilde{C}_D \chi^5 C^4 - C^B + \sqrt{\left(C - \frac{1}{4} K \tilde{C}_D \chi^5 C^4 - C^B \right)^2 + 4n_i^2} \right]. \quad (15)$$



(a)



(b)

Figure 4: Calculated concentrations of substitutionally dissolved phosphorus atoms (solid line), clustered phosphorus atoms (dash-dotted line), and electron density (dotted line) vs. the total dopant concentration for the formation of singly negatively charged clusters incorporating three (a) and four (b) phosphorus atoms. The experimental data are taken from Masetti *et al.* [12] for diffusion at 920°C .

It follows from the calculated dependence $n = n(C^T)$ that with an increase in the total phosphorus concentration C^T the concentration of charge carriers n reaches a maximum value $n_{\max} = 3.25 \times 10^8 \mu\text{m}^{-3}$ at $C_{\max}^T \sim 6.90 \times 10^8 \mu\text{m}^{-3}$ and then monotonically decreases. At maximum the concentration of substitutionally dissolved phosphorus atoms C_{\max} is 3.98×10^8

μm^{-3} . The fitting parameter $K\tilde{C}_D$ used in this calculation was equal to $1.9 \times 10^{-35} \mu\text{m}^9$.

For the case of doubly negatively charged clusters $(\text{DP}_4)^{2-}$ a disagreement is observed between the calculated dependence $n = n(C^T)$ and the experimental data. It follows from the comparison of the calculated dependences $n = n(C^T)$ with the experimental data that the suggestion on the formation of negatively charged clusters $(\text{DP}_2)^{2-}$, $(\text{DP}_3)^{2-}$, $(\text{DP}_3)^-$, and $(\text{DP}_4)^-$ provide a good fit of calculated functions to experimentally measured distribution of carrier concentration (see Figs. 3, 4). Therefore, it is difficult to extract the exact charge state and the number of the atoms incorporated into the cluster from the experimental data presented but the best fit is observed for the $(\text{DP}_4)^-$ clusters.

In Figs. 5a, 5b, 6a, and 6b the dependences $n = n(C^T)$ calculated for the clusters $(\text{DP}_3)^-$, $(\text{DP}_4)^-$, $(\text{DP}_3)^{2-}$, and $(\text{DP}_4)^{2-}$, respectively, are compared with the experimental data of [13] obtained for a temperature of 1000 °C. As can be see from the figures, all these clusters give a good fit to the experimental data, especially the $(\text{DP}_3)^{2-}$ and $(\text{DP}_4)^{2-}$ clusters. For the case of formation of the $(\text{DP}_3)^{2-}$ clusters, the system (10),(11) has the form

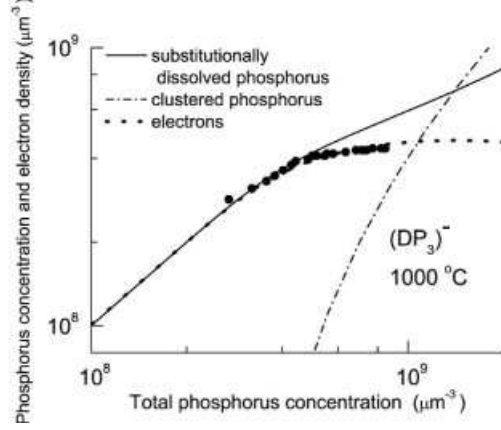
$$C^{AC} = K \tilde{C}_D \chi^5 C^3, \quad (16)$$

$$\chi = \frac{1}{2n_i} \left[C - \frac{2}{3} K \tilde{C}_D \chi^5 C^3 - C^B + \sqrt{\left(C - \frac{2}{3} K \tilde{C}_D \chi^5 C^3 - C^B \right)^2 + 4n_i^2} \right] \quad (17)$$

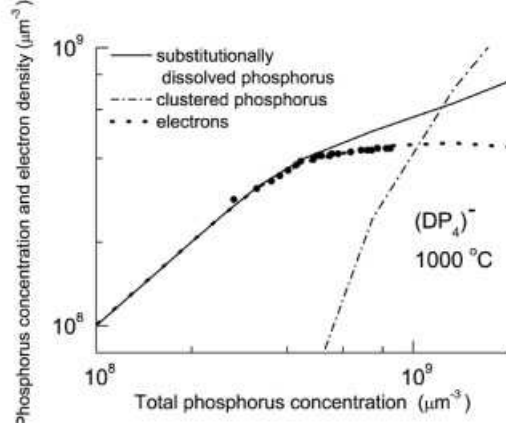
and the value of the fitting parameter $K\tilde{C}_D$ was chosen to be equal to $3.7 \times 10^{-27} \mu\text{m}^6$.

For the case of $(\text{DP}_4)^{2-}$ formation the system (10),(11) has the form

$$C^{AC} = K \tilde{C}_D \chi^6 C^4, \quad (18)$$



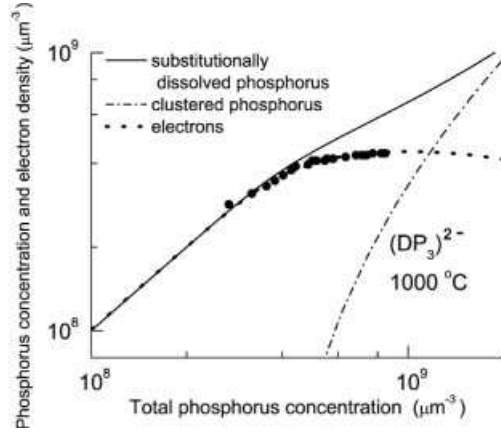
(a)



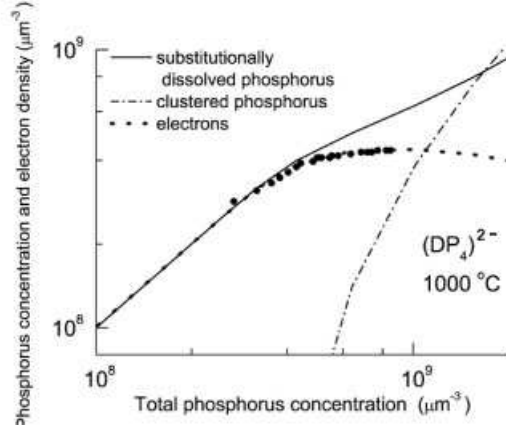
(b)

Figure 5: Calculated concentrations of substitutionally dissolved phosphorus atoms (solid line), clustered phosphorus atoms (dash-dotted line), and electron density (dotted line) vs. the total dopant concentration for the formation of singly negatively charged clusters incorporating three (a) and four (b) phosphorus atoms. The experimental data are taken from Solmi and Nobili [13] for diffusion at a temperature of 1000 °C.

$$\chi = \frac{1}{2n_i} \left[C - \frac{1}{2} K \tilde{C}_D \chi^6 C^4 - C^B + \sqrt{\left(C - \frac{1}{2} K \tilde{C}_D \chi^6 C^4 - C^B \right)^2 + 4n_i^2} \right] \quad (19)$$



(a)



(b)

Figure 6: Calculated concentrations of substitutionally dissolved phosphorus atoms (solid line), clustered phosphorus atoms (dash-dotted line), and electron density (dotted line) vs. the total dopant concentration for the formation of doubly negatively charged clusters incorporating three (a) and four (b) phosphorus atoms. The experimental data are taken from Solmi and Nobili [13] for diffusion at a temperature of 1000 °C.

and the fitting parameter $K\tilde{C}_D$ was chosen to be equal to $1.65 \times 10^{-37} \mu\text{m}^9$.

As for $(\text{DP}_2)^{2-}$ cluster formation, there is a little disagreement between the calculated electron density and experimental data. It is interesting to note that for the case of formation of doubly negatively charged clusters incorporating more than one phosphorus atom the values of parameters $K\tilde{C}_D$

providing a best fit to experimental data are equal for both temperatures 920 and 1000 °C. At the same time, the values of $K\tilde{C}_D$ calculated for singly negatively charged clusters are not equal for different temperatures. For example, if a formation of $(DP_4)^-$ is assumed, then the values of $K\tilde{C}_D$ are equal to 1.9×10^{-35} and $1.25 \times 10^{-35} \mu m^9$ for temperatures 920 and 1000 °C, respectively. It follows from the extracted values of $K\tilde{C}_D$ that the formation of doubly negatively charged clusters is most likely but it is difficult to choose between the clusters incorporating two, three or four phosphorus atoms.

3 Conclusions

Analysis of the models dealing with the dopant atom clustering in silicon was carried out. It is shown that the models of clustering developed do not explain the available experimental data, especially the effect of electron density saturation at high dopant concentrations. However, the phenomenon of saturation can be explained within the assumption of negatively charged phosphorus complex formation. To investigate this model, the electron density and concentration of dopant atoms incorporated into clusters are calculated for temperatures of 920 and 1000 °C as the functions of the total phosphorus concentration. A number of negatively charged clusters incorporating a point defect and phosphorus atoms, $(DP_1)^-$, $(DP_2)^-$, $(DP_2)^{2-}$, $(DP_3)^-$, $(DP_3)^{2-}$, $(DP_4)^-$, and $(DP_4)^{2-}$, are investigated. It is shown that in the case of $(DP_1)^-$ formation the concentration of charge carriers reaches saturation if the total phosphorus concentration increases. However, the saturation occurs only for very high values of the total dopant concentration that disagrees with experimental data. In the cases of clusters incorporating more than one phosphorus atom, the electron density reaches a maximum value and then decreases monotonically and slowly with an increase in the total dopant concentration. If a cluster incorporates 3-4 phosphorus atoms, the calculated dependences of carrier concentration agree well with the experimental data for both temperatures regardless of the value of the negative charge state of the cluster ($-$ or $2-$). On the other hand, for all doubly negatively charged clusters investigated the values of the fitting parameter are independent of the temperature. It means that formation of doubly negatively charged clusters is most likely but it is difficult to choose between clusters incorporating two, three or four phosphorus atoms. From a good agreement of the calculated curves with experimental data it follows that the model based on the formation of negatively charged clusters can be used in simulation of high concentration phosphorus diffusion.

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